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Differences in the Chemical Composition of Particulate Matter (PM) by their Size and their Importance in Ambient Sampling of PM

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1. Introduction

In regard to PM (particulate matter) air quality regulations, in many parts of the United States, and especially in areas that are (or have recently been) classified as in "non-attainment" for PM_{10} (2.5 to 10 μm in size) or $PM_{2.5}$ (less than 2.5 μm) such as the San Joaquin Valley (SJV), regulatory agencies are interested in contributions of agricultural sources to ambient concentrations of $PM_{2.5}$ and PM_{10}. With the removal of the permitting exemption from agriculture in 2003 as a result of California Senate Bill 700, agricultural industries in California have become a target of scrutiny. The SJV Air Pollution Control District has found that the available information on emission factors for agricultural operations is severely limited and needs improvement.

Several regulatory agencies have suggested using a characteristic percentage of one fraction of PM to estimate emissions of another fraction of PM. For example, in AP-42 (EPA's Compilation of Air Pollutant Emission Factors), $PM_{2.5}$ emissions from many sources are estimated as a fraction of the PM_{10} emission factors. Many of the PM_{10} emission factors in use today were developed by assuming that a static fraction of total suspended particulate (TSP) was PM_{10}. In the 1990's, in the absence of other data, it was common practice to assume that 50% of TSP was PM_{10}, as outlined by the use of this assumption in many California Air Resources Board (CARB) rules. Much like their approach to estimating PM_{10} emissions, CARB has also proposed using specified fractions of recorded TSP or PM_{10} emission factors to estimate $PM_{2.5}$ emission factors based on general PM emission profiles.

The chemical composition of $PM_{2.5}$ is often substantially different than that of coarse particles ($PM_{2.5}$ to PM_{10}), indicating that these particles originate from different sources. As a result, the use of a static fraction of TSP or PM_{10} emission factors to estimate $PM_{2.5}$ emissions will yield inaccurate estimates of emissions from many sources and will reduce the ability of regulatory agencies to effectively protect public health and welfare through strategic targeting of emissions reductions strategies.

The contents of the previous White Paper from the USDA-Agricultural Air Quality Task Force (AAQTF, 2009) discussed the significant over-sampling issues associated with the use of EPA Federal Reference Methods (FRMs, http://www.epa.gov/ttn/amtic/files/ambient/criteria/reference-equivalent-methods-list.pdf)) for sampling particulate matter (PM) at sites dominated by large particles such as near agricultural operations as well as the variations in particle

size distributions of PM emitted from various agricultural sources. In this paper we provide additional discussion of why reliable estimates of $PM_{2.5}$ emissions cannot be determined by applying static ratios to ambient measurements of TSP (Total Suspended Particulate Matter) or PM_{10} (composite samples of less than 10 μm that include $PM_{2.5}$ or less than2.5 μm) without consideration of source characteristics, including particle size distributions and the chemical composition of PM samples.

Measurement of the mass and characterization of the chemical composition of PM in the atmosphere is required because of its effects on: (1) human health through inhalation and respiration; (2) visibility; (3) climate and (4) environmental receptors such as natural resources. PM is produced by both natural and by human-made sources (http://cfpub.epa.gov/ncea/cfm/nceapubtopics.cfm?ActType=PublicationTopics). PM in the air exists as a solid or as a liquid. PM suspended in a gas phase as in the air is also known as an aerosol. In the present case, the discussion is directed to solid phase particles only.

Figures 1 and 2 show how PM in the atmosphere can be classified by its size into three broad categories: (a) Aitkin nuclei (less than 0.1 μm), (b) accumulation mode particles (0.1 to 2.5 μm) and (c) coarse-mode particles (2.5 to 10 μm) (Finlayson-Pitts and Pitts, 2000; Whitby, 1978). Collectively, Aitkin and accumulation mode particles are known as fine PM (e.g. $PM_{2.5}$). In addition, the term TSP (Total Suspended Particulate Matter) is applied to all particles less than 100 μm and was the first regulated form of PM. Note: fugitive dust describes PM from non-point sources, where as a point source is a single, identifiable local source such as a smoke stack.

With regard to regulations and human health effects, coarse particles are classified as "inhalable" (mostly deposited in the upper part of the respiratory tract or nasal system and easily ejected from the body, e.g., by sneezing) and fine particles are classified as "respirable" (mostly deposited in the pulmonary or lung system and persistent in the body). With regard to ecological effects, the primary pathway for the entry of PM into ecosystem components is through the soil and through surface and ground waters.
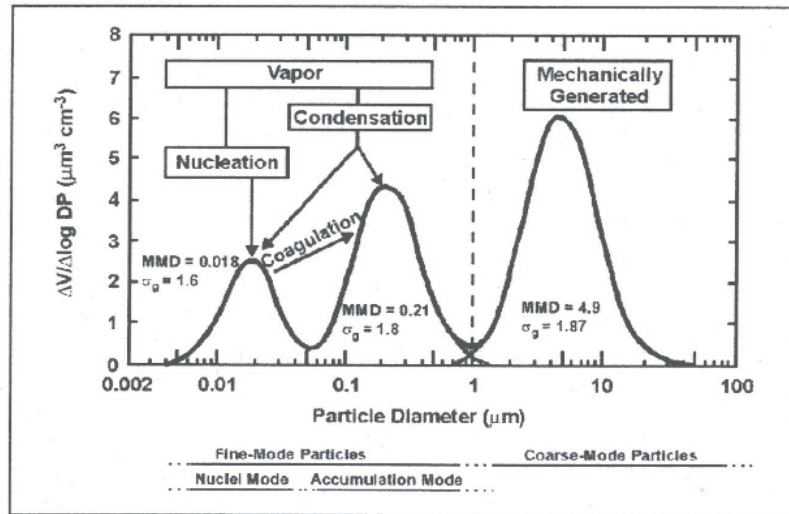
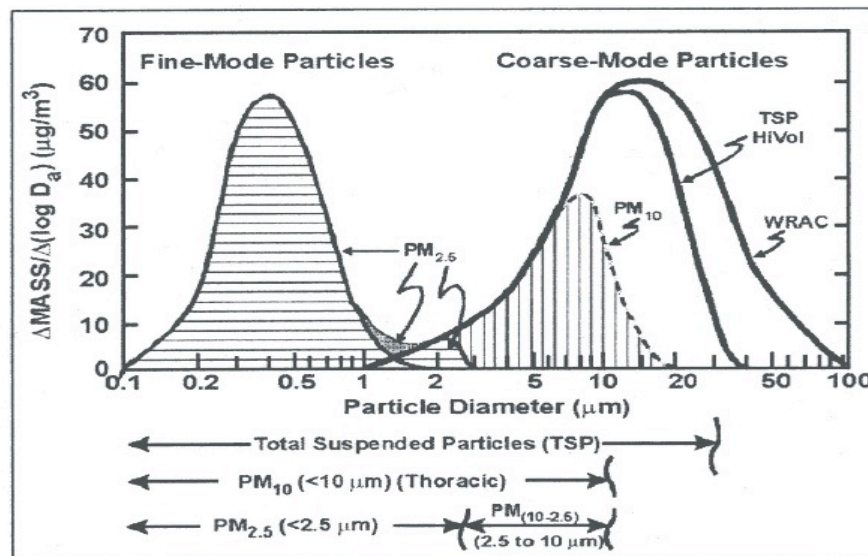**Figure 1. An idealized size distribution of ambient PM**

Figure 2. An idealized size distribution of ambient PM showing fine and coarse modes and the portions collected in various samples; HiVol : High Volume sampler; WRAC : Wide Area Aerosol Classifier(Lundgren and Burton, 1995). From Wilson and Suh, 1997.

2. Origins of TSP (less than100 μm), coarse or PM$_{10 – 2.5}$ and fine or PM$_{2.5}$ (less than 2.5 μm) particles

Fine or PM$_{2.5}$ can be either directly emitted to the atmosphere or formed by chemical reactions (i.e. secondary processes) in the atmosphere. To analyze the contribution of primary versus secondary aerosol contribution to total PM$_{2.5}$ on a regional basis, data from the Interagency Monitoring of PROtected Visual Environments (IMPROVE) project are being examined. The IMPROVE project measures PM$_{2.5}$, PM$_{10}$ and visibility at 156 National Parks and Wilderness areas across the US (most sites are rural but three are urban), and provides an analysis of

speciated fine or $PM_{2.5}$ concentrations in terms of ammonium sulfate, ammonium nitrate, organic and elemental carbon, and fine particle soil components. Ammonium sulfate and ammonium nitrate are formed by chemical reactions in the atmosphere involving sulfur dioxide (SO_2), oxides of nitrogen (NOx) and ammonia (NH_3). Organic carbon can be directly emitted (from processes such as fossil fuel combustion and biomass burning) and/or formed from trace gas reactions of volatile organic compounds (biogenic and anthropogenic sources) with atmospheric oxidants. Elemental carbon is largely a product of the combustion process, and along with fine soil particles, is directly emitted to the atmosphere. In the eastern US, ammonium sulfate and ammonium nitrate dominate the percent contribution to total $PM_{2.5}$, while in the western US, organic carbon (in many cases from fire) is the dominant contributor, with sulfates being the second highest. In the southwestern US there is a significant contribution of fine soil particles to total $PM_{2.5}$ where this is attributed to both Asian dust influences and local dust sources. Agriculture is not a source of many of these $PM_{2.5}$ precursor emissions.

In contrast, $PM_{10-2.5}$ coarse particles are directly emitted into the atmosphere (not formed by chemical reaction) from mechanical processes such as wind and abrasion of materials, and are crustal in nature. They can also be emitted from combustion processes. Particulate matter from agricultural operations is primarily composed of mechanically generated, crustal particles whose size distributions are characterized by large diameters and broad distributions (Table 1).

Table 1. Particle size distributions from various sources of PM.

Source	MMD[a] (µm AED)	GSD[b]	% $PM_{2.5}$	Reference
Urban	5.7	2.25	15	USEPA (1996)
Almond Sweeping	11.7	3.0	8	Faulkner and Capareda (2009)
Almond Pickup	12.3	2.6	5	Faulkner and Capareda (2009)
Cotton Harvest	14.3	2.2	1	Wanjura et al. (2009)
Dairy	15	2.1	1	Faulkner et al. (2007)
Feedyard	20	2.2	0.4	Faulkner et al. (2007)
Cotton Gin	23	1.8	0.01	Faulkner et al. (2007)
Broiler House	24	1.6	<0.01	Lacey et al. (2003)

[a] MMD = mass median diameter (expressed in aerodynamic equivalent diameter (AED))
[b] GSD = geometric standard deviation

TSP, by including all particles smaller 100 um in aerodynamic diameter, also contains $PM_{2.5}$ and $PM_{10-2.5}$, however, on a mass basis TSP is comprised of biological (pollen, spores, microorganisms, insects etc.), organic (e.g., persistent organic compounds such as certain pesticides), and inorganic (fugitive dust, sea spray, industrial dust etc.) species. Thus <u>TSP originates from a great diversity of sources that are highly variable in their size distribution and emission strength by location and in time.</u>

3. Physical properties of TSP (smaller than 100 μm), coarse or $PM_{10-2.5}$ and fine or $PM_{2.5}$ (smaller than 2.5 μm) particles

During dry conditions (periods of no precipitation), <u>both TSP and coarse particles are deposited on to surfaces by impaction or rapid gravitational settling (Davidson and Wu, 1990). Therefore, they do not travel long distances and are of local concern.</u> Under turbulent conditions, a portion of these large particles can be re-entrained into the atmosphere, travel some distance and be deposited again (somewhat analogous to hop, skip and jump). An exception, however, is the complex phenomenon of trans-continental transport of coarse particles (e.g., transport of Saharan dust from West Africa to Florida), a subject not discussed here.

In contrast to TSP and PM_{10}, a large percentage of <u>$PM_{2.5}$ are produced in the atmosphere in measurable concentrations at some distance from the sources of the precursor pollutants (e.g., sulfur dioxide gas being converted at a rate of ≤ 2% per hour in a coal-fired power plant plume to sulfate particles)</u> and are deposited on to surfaces by Brownian motion (the random motion of microscopic particles, suspended in a liquid or gas such as air, caused by collision with the molecules of the surrounding medium). Compared to gases and $PM_{10-2.5}$, $PM_{2.5}$ particles have the lowest rate or velocity of deposition on to surfaces (Davidson and Wu, 1990) and therefore, accumulate in their volume in the atmosphere and consequently are of a regional scale problem (Finlayson-Pitts and Pitts, 2000; Krupa, 1997). Accumulation of fine particles in the air leads to a reduction in visibility, an increase in cloud formation and also changes in our climate.

4. Relationships between PM_{10}, $PM_{10-2.5}$ and $PM_{2.5}$

Correlations of 24-h concentrations of PM (coarse + fine), PM coarse (2.5 to 10 μm) and PM fine (less than 2.5 μm) at the same site show that, in Philadelphia and St. Louis, <u>$PM_{2.5}$ is highly correlated with PM, but poorly correlated with $PM_{10-2.5}$</u> (Wilson and Suh, 1997). Similarly, among sites distributed across these urban areas, the site-to-site correlations of 24-h concentrations are high for $PM_{2.5}$ (homogeneity in their mass) but not for $PM_{10-2.5}$ (heterogeneity in their mass).

It should be noted that $PM_{10-2.5}$ are primary aerosols with a high deposition velocity, travelling relatively short distances at the surface, while $PM_{2.5}$ are largely secondary aerosols with a very low deposition velocity, travelling large, regional or even continental scale distances. Thus the atmospheric residence or lifetimes explain the observed spatial differences in the concentrations of the two size fractions by Wilson and Suh (1997). It also indicates that a true $PM_{2.5}$ measurement at a central monitor can serve as a better indicator of the community-wide air concentration of fine (homogeneity in their mass) than of coarse (heterogeneity in their mass) particles.

5. Chemical and toxicological properties of TSP (less than100 μm), coarse or PM$_{10 – 2.5}$ and fine or PM$_{2.5}$ (less than 2.5 μm) particles

PM size and mass are very important considerations in examining their behavior. An equally important consideration is their chemical composition, the concentrations of the individual constituents, pH, solubility and reactivity (Krupa, 1997). Clearly there are differences in the chemical composition of PM$_{10-2.5}$ and PM$_{2.5}$ and between urban, forested and agricultural sites (Table 2). Note that crustal or mechanically generated elements (aluminum, silica, calcium) predominate in the coarse fraction (PM$_{10-2.5}$) while sulfate, ammonium and elements such as zinc and lead and organic compounds emitted through specific industries or other human activities (e.g., semi-volatile organic compounds in agriculture) predominate in the fine particle (PM$_{2.5}$) fraction (Krupa et al., 1987; Pratt and Krupa, 1985; Stevens et al., 1984, Finlayson-Pitts and Pitts, 2000). It is also important to note that toxic metals such as mercury, arsenic, vanadium and cadmium exist in PM$_{2.5}$, more so than in PM$_{10-2.5}$. These features have critical implications in human and environmental toxicology.

Table 2. Average aerosol composition (ng m^{-3} or 10^{-9} g m^{-3}) for fine (less than 2.5 μm) and coarse (greater than 2.5 μm) particles at an urban[1], a forested[1] and an agricultural location[2].

Variable	Houstan [1,3] (Urban)		SmokeyMountains [1,3] (Forest)		Lamberton [2,4] (Agriculture)	
	Fine	*Coarse[5]*	*Fine*	*Coarse[5]*	*Fine*	*Coarse[5]*
Sulfate	16,700 ± 1,380	1,100 ± 200	12,000 ± 1,300	NA[6]	2,783 ± 763	370 ±119
Nitrate	250 ± 260	1,800 ± 260	300 ± 300	NA[6]	189 ± 29	638 ±174
Ammonium	4,300 ± 390	< 190	2,280 ± 390	NA	1,006 ± 414	16 ± 7
Aluminum	95 ± 60	1,400 ± 420	20 ± 18	195 ± 101	40 ± 8	357 ± 55
Silica	200 ± 60	3,800 ± 1,000	38 ± 10	580 ± 262	109 ± 21	1,500 ± 228
Calcium	150 ± 8	3,100 ± 160	16 ± 1	322 ± 73	52 ± 8	847 ±119
Zinc	102 ± 6	68 ± 5	9 ± 1	< 4	7 ±1	2 ±1
Lead	483 ± 23	127 ± 10	97 ± 5	14 ± 1	17 ±3	2 ±1

[1] Modified from Finlayson-Pitts and Pitts (1986) as reported from the original works of Dzubay et al. (1982) and Stevens et al. (1984). See Finlayson-Pitts and Pitts for the original references.
[2] Modified from Pratt and Krupa (1985)
[3] Samples collected during the daytime only
[4] Samples collected during the 24 hours
[5] Coarse particles represent 2.5 to 15 μm in size; fine particles represent less than 2.5 μm in size. The size cut off for coarse particles was changed to 2.5 to 10 μm by EPA during the mid 1980s
[6] NA = Not analyzed

The overall narrative on the differences in the origin, size and chemical composition of $PM_{10-2.5}$ and $PM_{2.5}$ underscores the critical issue that while the mass between the two fractions may be correlated if they are sampled individually at a common location, their chemical compositions can be very different and not related with each other in their elemental concentration ratios, (http://cfpub.epa.gov/ncea/cfm/nceapubtopics.cfm?ActType=PublicationTopics).

This is a very critical point, because if the true mass of PM is to be derived from $PM_{10-2.5}$, then a co-linearity (similar behavior) in the variability of the concentrations and chemical composition of $PM_{10-2.5}$ and $PM_{2.5}$ must be demonstrated at sufficient number of sites representing the geographic area such as the farmland.

6.   A need to accurately characterize the chemical composition of $PM_{2.5}$ and $PM_{10-2.5}$ in regard to source apportionment and receptor modeling in agriculture

According to Hopke (2009) source apportionment is the estimation of the contributions to the airborne pollutant concentrations that arise from the emissions of natural and anthropogenic sources. To obtain a source apportionment, data analysis tools called receptor models are applied to elicit information on the sources of air pollutants from the measured constituent concentrations. Typically, they use the chemical composition data for airborne PM samples. In such cases, the outcome is the identification of the pollution source types and estimates of the contribution of each source type to the observed concentrations. The composition of $PM_{2.5}$ and $PM_{10-2.5}$ is important to source apportionment and receptor modeling illustrating the need for accurate source profiles so that the results can be useful to the regulatory agencies that rely heavily on the modeling. If such modeling is done with inaccurate data, then inappropriate regulatory decisions may be made as previously pointed out by the Agricultural Air Quality Task Force (AAQTF).

7.   Artifacts introduced in the data on the chemical composition of $PM_{2.5}$ through the use of TSP and PM (coarse + fine) measurements

The fine particle or $PM_{2.5}$ mode typically includes most of the total number of particles in the atmosphere and a large fraction of the mass for example, about one third of the total mass in non-urban areas and about one half in the urban areas. In that context, it is very important to note that both positive and negative errors (biases) occur during the sampling of PM. The gain (positive bias) is due to the conversion of gas phase molecules to particles on the sampler filter or the loss (negative bias) is due to the volatilization of compounds from the particle phase to the gas phase on the sampler filter (e.g., ammonium nitrate near feed lots; semi-volatile organic compounds such as pesticides). Further, cross-contamination can occur during sampling between particles smaller than 2.5 µm and particles larger than 2.5 µm. In addition to those effects, even where $PM_{10-2.5}$ and $PM_{2.5}$ are

sampled individually as with a dichotomous sampler, cross contamination of about 5% can occur between the particles larger than 2.5 μm and particles smaller than 2.5 μm in terms of their sulfate, nitrate and inorganic elemental concentrations (which can be used as tracers to examine the cross contamination).

One approach to estimating the cross contamination between PM larger than 2.5 μm and PM smaller than 2.5 μm chemical composition (an indicator of sampling efficiency and accuracy) is to use an enrichment factor. Here Al (Aluminum), Ca (Calcium) and Si (Silica) are considered to be indicative of crustal or coarse particles. Thus for example, since Al from the soil (crust) has the least variability at a given location, the enrichment factor (EF) for a particular element "X" can be defined as:

$$EF_{crust} = \{X_{air}/Al_{air}\} / \{X_{crust}/Al_{crust}\}$$

Thus, crustal materials such as calcium that predominate in the coarse (greater than 2.5 μm) fraction will have an average EF value close to unity (1.0), while another element such as copper will have an average value of 100 (Milford and Davidson, 1985). Therefore, changes in EF values in the fine particle fraction (lesser than 2.5 μm) can be used to calculate the level of cross contamination by the coarse particles (2.5 to 10 μm) during sampling in support of the discussion in the previous White Paper on PM sampling artifacts (AAQTF, 2009). Here, an important characteristic that can be used is, in general particles larger than 2.5 μm are generally basic and particles smaller than 2.5 μm are generally acidic in their nature. However, EF factors (atmosphere versus crustal) can not be used in the present context unless discreet measurements are taken of true PM $_{10 - 2.5}$ and PM$_{2.5}$ at sufficient number of locations representative of both urban and non-urban locations in a given geographic region such as the San Joaquin Valley, CA where agriculture is predominant.

8. Conclusions

Table 3 (below) provides a summary of the general differences in the characteristics of PM$_{10 – 2.5}$ and PM$_{2.5}$.

(a) There are significant differences in the origin (primary versus secondary aerosols), their atmospheric residence times, transport, and rates of dry deposition and geographic distribution of coarse or PM$_{10-2.5}$ and fine (less than 2.5 μm) particles.

 (b) Correlations of 24-h concentrations of PM (coarse + fine), PM coarse (2.5 to 10 μm) and PM fine (less than 2.5 μm) at the same site show that PM $_{2.5}$ is highly correlated with PM, but poorly correlated with PM $_{10 -2.5}$ Further, where PM$_{10- 2.5}$ (coarse only) and PM $_{2.5}$ are sampled individually (e.g., with a dichotomous sampler) at a common location and sampling period, both their mass and chemical

compositions will be very different and not related with each other in their elemental concentrations and their ratios and consequently differences in human and environmental effects.

(c) Among sites distributed across urban areas, the site-to-site correlations of 24-h concentrations will be high for PM $_{2.5}$ but not for $PM_{10-2.5}$ or PM_{10} (coarse + fine). This adds to the concern with using TSP or PM_{10} (any measure with coarse PM) as a surrogate for $PM_{2.5}$ as the resulting $PM_{2.5}$ measure will be highly influenced by distance to coarse PM Sources.

(d) In the context of regional scale PM monitoring, if the true concentrations of PM $_{2.5}$ are to be derived from ambient PM_{10} concentrations, then a co-linearity (similar behavior) in the concentrations of coarse (PM $_{10-2.5}$) and fine (less than 2.5 μm) particles must be demonstrated.
That can be achieved by examining the chemical composition of the <u>individual size fractions</u> at sufficient number of locations representing the geographic area. Possible changes in alkalinity and acidity respectively of coarse and fine particle samples can be used as a preliminary screening tool for identifying cross contamination during sampling between $PM_{10-2.5}$ and $PM_{2.5}$.

(e) In the final analysis, while discreet samples of PM $_{10-2.5}$ and $PM_{2.5}$ (such as with the use of a dichotomous sampler) can be correlated, arithmetic calculation of $PM_{10-2.5}$ cannot be well correlated with $PM_{2.5}$ per se (Wilson and Suh, 1997).

(f) Furthermore discreet measurements of the chemical composition of fine (less than 2.5 μm) and coarse (2.5 to 10 μm) particles are required to apportion the contribution of specific sources to the general air quality and to the implementation of regulatory policies relevant to agriculture (primarily sources close to the ground level).

9.   Recommendations

(a) To address the three issues outlined below, we recommend the establishment of an appropriate pilot agricultural PM monitoring network that represents the five major crops in the US, corresponding soil factors, climate zones and cropping practices (e.g., tillage, irrigation).

Such an effort could be coupled to other ongoing programs that address agriculture related PM. A future consideration in the overall PM monitoring program should be the inclusion of characterizing the role of animal agriculture.

(b) To obtain the true concentrations of PM $_{2.5}$ from PM_{10} values, we recommend that a co-linearity (similarity in behavior) in the concentrations and elemental ratios of coarse (2.5 to 10 μm) and fine (less than 2.5 μm) particles be demonstrated at sufficient number of locations representative of the agricultural region under consideration.

 (c) We recommend that discreet measurements of the chemical composition of fine (less than 2.5 μm) and coarse (2.5 to 10 μm) particles be made to apportion the contribution of specific sources to the general air quality and to the implementation of regulatory policies germane to agriculture.

(d) In regard to recommendation (c) we strongly support the need to understand the conditions and source strengths that govern exceedences of PM air quality regulatory standards. For example, wood burning during the cold season or fugitive dust during the dry season.

10. References

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Table 3. General differences observed in the characteristics of coarse and fine particles. Both particle fractions are included in TSP.

Characteristic	Coarse	Fine
Origin	Primary aerosol, produced directly by a source	Secondary aerosol, produced mainly by reactions in the atmosphere
Geographic distribution	Mainly local	Regional, continental
Size	2.5 to 10 µm	Smaller than 2.5 µm
Deposition (during dry periods)	Gravitational settling	Brownian motion
Dry deposition velocity or rate	Relatively very high	Relatively very low
Atmospheric lifetime	Mainly minutes to hours	Days to weeks
Concentrations in a region	Very heterogeneous	Relatively homogeneous
Human uptake	Inhalable particles	Respirable particles
Chemical composition	Primarily crustal elements such as aluminum, calcium, silica	Primarily sulfate, nitrate, ammonium, trace metals such as arsenic, beryllium, cadmium, mercury, vanadium
Solubility	Largely insoluble Not –deliquescent	Largely soluble, Deliquescent